



Green zero valent iron nanoparticles dispersion through a sandy column using different injection sequences



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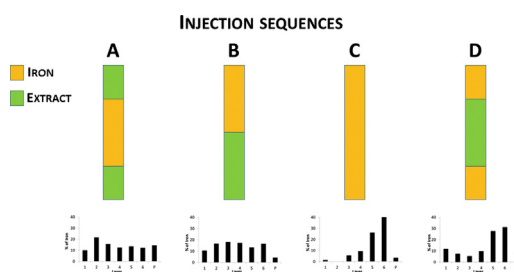
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HIGHLIGHTS

- nZVI transport is influenced by soil particle size and saturation level.
- The injection sequence for nZVI synthesis might change nZVI mobility trend.
- Saturated tests led generally to lower mobility nZVI rates.
- Smaller sand sizes hinder nZVI transport.
- Depending on the contamination type, several strategies with nZVI may be applied.

GRAPHICAL ABSTRACT



ARTICLE INFO

Article history:

Received 14 March 2018

Received in revised form 7 May 2018

Accepted 7 May 2018

Available online xxxxx

Editor: D. Barcelo

Keywords:

Nanoparticles

nZVI

Green synthesis

Dispersion

Mobility

Soil contamination

ABSTRACT

The contamination of soils is a global environmental problem that urges an increased effort to recover polluted sites. In Europe, there are an estimated 20,000 polluted sites that need to be remediated and around 350,000 sites that are classified as potentially contaminated by the European Environment Agency (EEA). Remediation is a part of the solution to this problem, requiring the most innovative and sustainable technologies. In this context, the use of zero valent iron nanoparticles (nZVI) is a promising, low cost and efficient technology for the remediation of soil and groundwater contaminated with a wide range of organic and inorganic pollutants. Among the nZVIs, the ones produced using Green synthesis methods (green nZVIs (gnZVI)) using natural extracts, such as green tea, are increasingly considered an alternative technology for the future. However, there are issues related to the application of gnZVI in soil that are still not fully understood, requiring further research, among these is the study of their transport in soils.

Therefore, this work aims to study the transport of gnZVIs in sandy soils under diverse conditions such as soil particle size, soil saturation level and injection sequence. Several experiments were performed in an acrylic column with two sandy soils with different particle sizes (between 0.5 and 1.0 mm and higher than 1.0 mm), using two distinct water saturation conditions (saturated and dry) and four injection sequences. After these tests the distribution of the gnZVI along the soil column was determined by atomic absorption spectroscopy. This work allowed concluding that the injection sequence might be one of the most important factors influencing the rate of nZVI dispersion through a sandy column. According to the results it was possible to propose, for distinct types of contamination (deep, superficial, hot spot or dispersed), the most appropriate injection sequence.

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1. Introduction

Nanoparticles (NPs) are defined as any particle that has at least one dimension between 1 and 100 nm (Christian et al., 2008). These

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materials can be classified according to their origin: natural (biogenic, geogenic, atmospheric or pyrogenic) or man-made materials (engineered or unintentionally produced as by-products) (Matlochova et al., 2013); or according to the NP composition: inorganic or carbon-based (Tratnyek and Johnson, 2006; Nowack and Bucheli, 2007). Nowadays, NPs are used in a wide range of applications such as electronics, medicine, pharmaceutical products and environmental applications. Among these materials, metallic iron NPs are widely accepted by many users and regulatory agencies because of their low production costs and high reactivity (Zhang, 2003). Over the last years, extensive laboratory studies have demonstrated that zero valent iron nanoparticles (nZVI) can be used to decompose or stabilize several and distinct contaminants such as halogenated hydrocarbons (Zhang, 2003; Nurmi et al., 2005), polychlorinated biphenyls (Varanasi et al., 2007), organochlorine pesticides (Elliott et al., 2003), organic dyes (Liu et al., 2005), various inorganic compounds (Cao et al., 2005), and distinct metal ions (Li et al., 2017).

nZVI can be synthesized through bottom-up or top-down methods. The green bottom-up method is the one that is becoming more attractive and is gathering the attention of the scientific community (Machado et al., 2013). The Green synthesis is based on the use of plant extracts or other natural products or wastes (such as wastes from the juice industry, namely citrines peel, albedo and pulps) which promote the reduction of iron salts and the subsequent formation of nZVI (Machado et al., 2014). Husen and Siddiqi (2014) reported that extracts from almost all plants (herbs, shrubs or trees) that contain aromas, flavonoids, phenols, alcohols and proteins can produce metal NPs when reacting with the correspondent metal salt. The Green synthesis is environmentally friendly because a toxic reagent (sodium borohydride) is replaced by a natural extract, that in this case is inexpensive (Machado et al., 2013), and its procedure is simple and easy to implement on a large scale (Machado et al., 2014).

The use of the Green synthesis was first applied by VeruTEK and USEPA (Hoag et al., 2009) using plant extracts (coffee, green tea, black tea, lemon, balm, sorghum, bran, grape etc.) with high antioxidant capacities, mainly because of the presence of polyphenols, that react with iron (II) solution, producing nZVI. Machado et al. (2013) concluded that the production of nZVI can be improved using plant leaves with low moisture contents. On the other hand, Mystrioti et al. (2015) and Kozma et al. (2016) observed the formation of other forms of iron (namely oxides or hydroxides) that can lead to a higher aggregation, resulting in a lack of mobility in porous systems (Huang et al., 2014).

The success of a remediation in which injected nZVI are used depends on their mobility in the porous media, the contact between the nZVI and the contaminant, its reactivity and the impact of the material on the contaminated ecosystem (Husen and Siddiqi, 2014). If nZVI is exposed to air or water, several processes, including break-downs and auto-reduction within the oxide shell can occur (Hoerle et al., 2004), in the case of aqueous systems nZVI is oxidized to Fe^{2+} (fast process) and/or Fe^{3+} (slower process) (Tosco et al., 2014). These phenomena result in a loss of zero valent iron content (Liu and Lowry, 2006; Sarathy et al., 2008), decreasing the capacity to react with a target contaminant. In addition, the nZVI particles may aggregate and a new, mixed-valence ($\text{Fe}^{2+}/\text{Fe}^{3+}$) oxide layer (Sarathy et al., 2008) can be formed. According to Zhao et al. (2016) several factors affect the size and stability of nZVI, including: temperature, natural organic matter, pH, ionic strength and type and concentration of stabilizers (He and Zhao, 2008; Zhang et al., 2011). Different conditions are proven to affect the reactivity of nZVI namely the surface area (Nurmi et al., 2005), the age of the NP (Liu and Lowry, 2006), the pH of the aqueous phase (Song and Carraway, 2005; Liu and Lowry, 2006), the use of stabilizers (He and Zhao, 2005; He et al., 2010; Sakulchaicharoen et al., 2010) and the concentrations of the contaminants and other reactive groundwater constituents (Liu et al., 2005; Song and Carraway, 2005).

In the last decade, several field tests using nZVI have been performed to evaluate their efficiency in degrading distinct contaminants and to

understand their mobility along the soil matrix (Quinn et al., 2005). However, in several of these field tests poor nZVI mobility was observed, probably due to several interactive forces between the NPs and the soil particles and due to soil pore blockage because of nZVI agglomeration (Quinn et al., 2005; Bennett et al., 2010). Even in laboratory experiments conditions nZVI tends to aggregate, producing clusters that can reach colloidal sizes (several micrometers) that limits their remediation potential (Ryan and Elimelech, 1996; Kharisov et al., 2012). Without the application of surface coatings to aid colloidal stability, rapid aqueous aggregation of standard nZVI is frequently observed (Schrick et al., 2004). When the size of the NPs is similar or larger than the soil pores, they become trapped, thus the transport will likely be reduced by straining, filtration or sedimentation by the establishment of interactions between the soil-water and soil matrix (Nowack and Bucheli, 2007; Fang et al., 2009). Consequently larger aggregates might be retained in the upper soil layers (Fang et al., 2009). The retention mechanisms in the unsaturated zone are even more complex than in the saturated zone, mainly due to the presence of an air phase in the system (Bradford and Torkzaban, 2008). In the former, water flow is restricted by capillary forces to the smaller regions of the pore space and flow rates are relatively small (Bradford and Torkzaban, 2008), where the transport may be influenced by increased attachment to the solid-water interface (Chu et al., 2001), attachment to the air-water interface (Cherrey et al., 2003), film straining in water films enveloping the solid phase (Saiers and Lenhart, 2003), and retention at the solid-air-water triple point (Steenhuis et al., 2006). Moreover, transients in water content during infiltration and drainage processes can also play a significant role (Saiers and Lenhart, 2003).

Bennett et al. (2010) concluded that the mobility of the nZVI decreased over time, making it necessary to pump the groundwater to force the nZVI towards the contaminated hot zone. Quinn et al. (2005) also observed that emulsified nZVI were not evenly distributed within the target treatment area and did not percolated as far as it was previously expected. Following this, He and Zhao (2005) stated that the soil mobility or deliverability is the most prominent technical obstacle of the application of nZVI in soils.

The knowledge of the mobility of NPs in porous media and the processes that affect their movement is limited, especially for unsaturated ones, mostly due to the complex nature of the interactions between the NPs and the soil matrix (Tiede et al., 2009; Tourinho et al., 2012). In order to understand the mobility of nZVI in sandy soils and the factors that affect their movement, this work aimed to study the influence of moisture content, particle size and the green nZVI injection sequence on the dispersion of the nZVI along a sandy soil column.

This paper describes the first work of a wider project concerning the study of the transport of gnZVI in distinct types of soils, evaluating the impact of several operational (sequence and location of injection) and physico-chemical (such as clay, sand and organic matter contents) parameters on the gnZVI distribution in soils. This first work focused on the study of the transport of gnZVIs in sandy soils where different injection sequences were tested.

2. Materials & methods

2.1. Reagents and solutions

Iron (III) chloride was obtained from Merck. Type II deionized water (resistivity = 15.0 M Ω cm) was used throughout the study and was obtained from an Elix 3 Advantage system (Millipore). Black tea (Tetley) was used to produce the extracts. A 0.1 M iron (III) chloride solution was prepared and used to synthesize nZVI.

2.2. Analysis of iron

The quantification of the iron in the soil samples was performed using an Analytik Jena CONTRAA 700 atomic absorption spectrometer.

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